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Nonionic surfactant Brij35 effects on toluene biodegradation in a composite bead biofilter

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Nonionic surfactant effects on the toluene dissolved in the water phase and biodegradation kinetic behaviors of toluene in a composite bead biofilter were investigated. The toluene dissolved in the water phase was enhanced by the addition of surfactant into aqueous solution and the enhancing effect was more pronounced in the surfactant concentration less than critical micelle concentration. For the microbial growth process, the microbial growth rate was inhibited at higher surfactant content and the degree of inhibitive effect was more pronounced at lower inlet concentration. The microbial growth rate was inhibited at higher inlet concentration. Zero-order kinetic with the diffusion limitation could be regarded as the most adequate biochemical reaction model. For the biochemical reaction process, the biochemical reaction rate was also inhibited at higher surfactant content and the degree of inhibitive effect was more pronounced at lower inlet concentration. The biochemical reaction rate was also inhibited at higher inlet concentration. The maximum elimination capacity decreased with increasing surfactant content and it was in the range of 17.41 to 26.12 g C h^{-1} m^{-3} bed volume. The addition of surfactant into filter material was unfavorable for toluene degraded by the microbial.

Key words: Nonionic surfactant, toluene, biodegradation, composite bead biofilter.

INTRODUCTION

The removal of volatile organic compounds (VOCs) from a polluted air stream using a biological process is highly efficient and has low installation and operation maintenance costs. Biofiltration technology offers environmental advantages; it does not generate undesirable by-products by converting many organic and inorganic compounds into harmless oxidation products (water and carbon dioxide). Biofiltration involves the passage of a polluted air stream through a packed bed containing microorganisms immobilized within a biofilm attached to the bed-packing material. Contaminants are transferred to the interface between the gas and biofilm and are subsequently absorbed into the biofilm. Contaminants are then used as carbon and/or energy sources for the microorganisms within the biofilm. The solid filter material provides a nutrient source and matrix for the attachment of microorganisms in the biofiltration process. Therefore, the filter material property is an important factor in

obtaining optimal pollutant removal. The optimal filter material should have the following characteristics: high moisture holding capacity, porosity, available nutrients and pH buffer capacity (Deviney et al., 1999). A spherical polyvinyl alcohol (PVA)/peat/KNO_3/GAC composite bead was prepared and proved to be suitable as a filter material in the biofiltration process in our previous works (Chan and Lin, 2006; Chan and Peng, 2008).

Toluene is widely used industrial chemicals. Toluene is a hydrophobic compound and one of the 189 hazardous air pollutants listed in the 1990 Clean Air Act Amendment (CAAA90) proposed by the US Environmental Protection Agency (EPA). Even at low concentrations, toluene has been found to be carcinogenic, cause damage to the liver and kidney and paralyze the central nervous system (Martin et al., 1998; Murata et al., 1999). Thus, large volumes of toluene are released into the atmosphere during manufacturing process every year, endangering the air quality and public health. The use of surfactants has the potential to increase the biodegradation rate of hydrophobic organic compounds in contaminated environments by increasing the total aqueous solubility of these compounds (Tiehm, 1994; Volkering et al., 1995;

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Grimberg and Aitken, 1995; Meeren and Verstratete, 1996; Guha and Jaffe, 1996). Nonionic surfactants are usually used in the bioavailability studies due to their relatively low toxicity compared to ionic surfactants (Yeh et al., 1998). Brij 30 was the most biodegradable surfactant among Brij30, Tween 80 and Trition X-100 three nonionic surfactants, and the solubility of naphthalene and phenanthrene in the water phase was linearly proportional to the concentration of nonionic surfactant (Kim et al., 2001). The biodegradation rate of pentachlorophenol (PCP) was enhanced for the concentration of PCP at 140 and 220 mg L^{-1} as the concentration of nonionic surfactant Tergitol NP-10 increased from 0 to 1500 mg L^{-1}, but it was inhibited for the concentration of PCP at 100 and 50 mg L^{-1} (Cort et al., 2002). Sodium dodecyl sulfate (SDS) anionic surfactant was found to inhibit growth of the fungus and polyoxyethylene sorbition monolaurate (Tween 20) nonionic surfactant was found to enhance inoculums development by shortening the lag period and toluene degradation in fungal vaporphase bioreactor (Woertz and Kinney, 2004). The removal efficiency of gaseous trichloroethylene and tetrachloro-ethylene was enhanced respectively, when a nonionic surfactant Alfonic[R] 810-60 was introduced into activated carbon biofilter (Kim et al., 1999).

The filter material containing surfactants would enhance the performance of biofilter treating hydrophobic containments. But a prepared filter material containing surfactant in the biofiltration process is unavailable in the relevant literature. We had obtained that the process for degradation of VOCs in a composite bead biofilter could be divided into lag phase, exponential phase and stationnary growth phase, with the exponential and stationary growth phases which are important for controlling the removal efficiency of biofilter (Chan and Lin, 2006; Chan and Peng, 2008). Therefore, studies of the kinetic of exponential growth and stationary phases were very important for the operation and design of the biofiltration process. However, details of the kinetic of such biodegradation process in biofilter are scant in the relevant literature. This article investigates the preparation of a spherical PVA/peat/KNO$_3$/GAC composite bead contained non-ionic surfactant Brij 35 and bio-degradation kinetic behaviors of hydrophobic toluene compounds in this composite bead biofilter. The effect of inlet concentration and surfactant content in this biofilter on the microbial growth rate and biochemical reaction rate are also studied.

MATERIALS AND METHODS

Materials

Peat (industrial grade from KekkilaOyj, Tuusula, Finland) was dried at 105°C before use. It has a dry density of 90 kg m^{-3}, a pH of 5.5, a pore volume of 96% and an organic substance content of 91%. Boric acid, sodium monobasic phosphate, sodium dibasic phosphate, potassium nitrate, and toluene (extra pure grade from Union

Chemical, Hsinchu, Taiwan) were used as received. Poly(vinyl alcohol) (PVA) powder (industrial grade from Chung Chun Petrochemical, Hsinchu, Taiwan), granular activated carbon (GAC) (industrial grade from Taipei Chemical, Hsinchu, Taiwan) and Brij 35 (industrial grade from Imperial Chemical Industries Limited, London, England) were also used as received. The Brij 35 has a molecular formula of $C_{12}H_{25}(OCH_2CH_2)_{23}OH$, a hydrophilic-lipophilic balance (HLB) of 16.9 and a critical micelle concentration (CMC) in the range of 70 to 110 mg L^{-1} (Yeh et al., 1998; Zhou and Zhu, 2004; Laha and Luthy, 1992; Park and Pritchard, 2002)

Dissolution experiments

The experiment of toluene dissolved in the water phase was carried out by adding 3.0 g toluene into a desired surfactant concentration aqueous solution (150 ml) in a glass-stopper Erlenmeyer 250 ml flask. The desired surfactant concentration was ranging from 0 to 540 mg surfactant L^{-1}. The flasks were in a shaking isothermal water bath for about 24 h at 30°C. Then, the solution was centrifuged for 1 h at 2000 rpm to separate into water and organic phases. The amount of toluene dissolved in the water phase was measured by gas chromatography (GC) (Model GC-8A from Shimadzu, Tokyo, Japan).

Biofilter experiments

The procedures for preparing PVA/peat/GAC/KNO$_3$ composite beads and the apparatus and operation of the biofilter system were described in our previous work (Chan and Chang, 2006; Chan and Peng, 2008). Before packing, the prepared PVA/peat/GAC/KNO$_3$ composite bead was immersed in a 0.384 M KNO$_3$ and a desired surfactant concentration aqueous solution to adsorb KNO$_3$ and surfactant, and to reach equilibrium (approximately 24 h). The bead moisture content was humidified to more than 1.5 g water g^{-1} dry composite bead and the seeding was performed with activated sludge obtained from the wastewater treatment plant in Hsinchu Science-Based Industry Park. As the stationary phase had been maintained for more than 3 d, the biofilter operating was stopped according to the variations of toluene removal efficiency with operation time. Then, new filter material was repacked and the operation procedures described above were carried out to start another experiment with the desired inlet concentration and surfactant content. The gas flow rate was maintained at 0.03 m^3 h^{-1} for all experiments and consequently the empty bed residence time (EBRT) of biofilter column was 96.5 s. The toluene concentration in the inlet and exit air streams of each section was determined by auto-sampling and analyzed using gas chromatography (GC) (Model GC-8A from Shimadzu, Tokyo, Japan). There was no product detected except CO$_2$ and water in the exit air stream. The toluene removal efficiency was calculated by the difference of the toluene concentration between the inlet and exit air streams. The relative standard deviation and relative error of the experimental measurements were less than 2 and 5% respectively.

RESULTS AND DISCUSSION

Dissolution capacity

The variations of S/S$_0$ value with surfactant concentration are shown in Figure 1. S/S$_0$ was the amount of toluene dissolved in the water phase of the aqueous solution with and without surfactant, respectively. It was found that all the S/S$_0$ value were greater than 1.0 in the surfactant
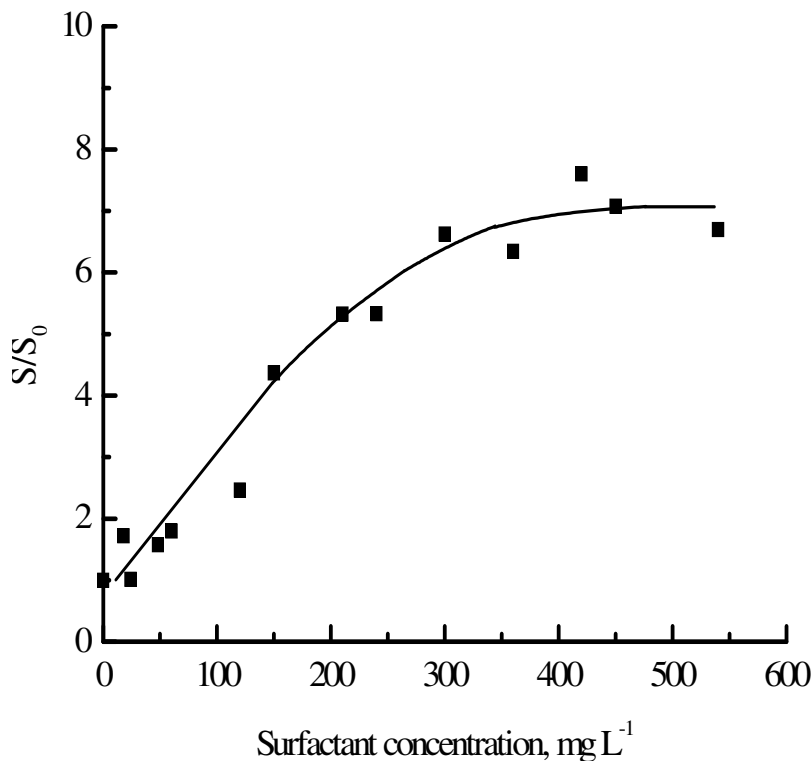
Figure 1. The variations of S/S$_0$ value with surfactant concentration.

concentration range of 0 to 540 mg L^{-1}. The result indicated that the addition of surfactant into aqueous solution would enhance the toluene dissolved in the water phase. The S/S$_0$ value sharply increased with increasing the surfactant concentration in the concentration range of 0 to 150 mg surfactant L^{-1} and then gradually to reach an equilibrium value (about 6.87) in the concentration range of 300 to 540 mg L^{-1}. The result indicated that the enhancing effect in the surfactant concentration less than CMC was more pronounced than that in the surfactant concentration higher than CMC.

The CMC of Brij 35 in the range of 70 to 110 mg L^{-1} and the enhancing effect for the toluene dissolved in the water phase was more pronounced in the Brij 35 concentration less than CMC. In order to study whether the biodegradation of toluene was enhanced in the surfactant concentration less than CMC or not, we chooses three surfactant concentrations as the desired surfactant concentration of aqueous solution for the composite bead immersed in and adsorbed surfactant for the later experiments. Three desired surfactant concentrations were 50 mg L^{-1} (which was about half of CMC and lies in the S/S$_0$ value sharply increased region), 210 mg L^{-1} (which was about double of CMC and lies in the S/S$_0$ value gradually increased region) and 420 mg L^{-1} (which lies in the S/S$_0$ value at an equilibrium value region). Therefore, the calculated surfactant content in the composite bead was 0, 0.054, 0.225 and 0.459 mg g^{-1} composite bead for the surfactant concentration 0, 50, 210 and 420 mg L^{-1}, respectively.

Microbial growth process

The variations of toluene removal efficiency with operation time at various inlet concentration and surfactant content are shown in Figure 2. (Only the inlet concentration of 100 ppm and the surfactant content of 0.225 mg surfactant g^{-1} composite bead are shown). It was found that the variations of toluene removal efficiency with operation time appeared in three phases: lag phase (Phase I), exponential growth phase (Phase II) and stationary phase (Phase III) (Chan and Lin, 2006; Chan and Peng, 2008). Only the biochemical kinetic behaviors in the exponential growth phase and stationary phase was studied in this work. The elimination capacity of toluene was also determined.

In the exponential growth phase (Phase II), the microbial growth rate increased exponentially and was represented by the following equation (Valsaraj, 1995)

$$dX/dt = k_g\, X \tag{1}$$

Where x is the number of viable cells per unit volume, k$_g$ is the microbial growth rate and t is the operation time. The amount of contaminant degraded was proportional to the amount of viable cell produced in the biofilter because the kinetic of contaminant degradation is closely related to the kinetics of microbial growth (Valsaraj, 1995). Therefore, the concentration of VOCs in the exit air stream (C)
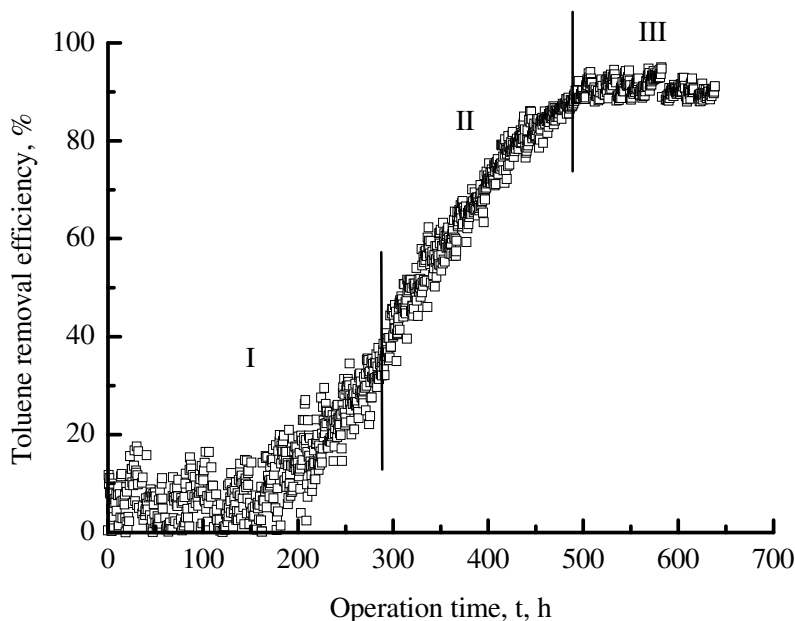
Figure 2. The variations of toluene removal efficiency with operation time at inlet concentration of 100 ppm and surfactant content of 0.225 mg g^{-1} composite bead.

was inversely proportional to the number of viable cells per unit volume in the bed, and Eq. 1 can be converted into

$$dC/dt = - k_g C \qquad (2)$$

Integration of Eq. 2 yields

$$\ln(C/C_0) = - k_g t \qquad (3)$$

Where C_0 is the concentration of VOCs in the inlet air stream. A plot of $\ln(C/C_0)$ versus t should correspond to a straight line and k_g can be determined. Therefore, the microbial growth rate k_g at various inlet and surfactant concentrations was calculated from the data in Phase II and Eq. 3.

The variations of the k_g values with surfactant content for four inlet concentrations are shown in Figure 3. The k_g value decreased with increasing surfactant content in the surfactant content range of 0 to 0.459 mg g^{-1} composite bead. The result indicated that the microbial growth rate was inhibited at higher surfactant content and the addition of surfactant into composite bead was unfavorable for microbial growth. The sequestration of a hydrophobic compound within surfactant micelles might decrease its biodegradation rate by decreasing its concentration in the water phase (Roch and Alexander, 1995). In addition, surfactants might produce toxicity in and decrease the activity of microbial (Volkering et al., 1995). Therefore, the

greater amounts of toluene dissolved into micelles in the biofilm or the greater toxicity in microbial at higher surfactant content lead to the greater inhibition on the biodegradation of toluene. The slope of the linear profiles in this surfactant content range for 50, 100, 200 and 300 ppm inlet concentration was 4.90×10^{-2}, 2.81×10^{-2}, 1.44×10^{-2} and 1.05×10^{-2} h^{-1} mg^{-1} g composite bead, respectively. The result indicated that the degree of inhibitive effect was more pronounced at lower inlet concentration. The k_g value in the inlet concentration range of 50 to 100 ppm was much higher than that in the inlet concentration range of 200 to 300 ppm. The result indicated that such an amount of toluene dissolved in the biofilm produced toxic to the microorganism in the inlet concentration range of 200 to 300 ppm. The k_g value decreased with increasing inlet concentration in the inlet concentration range of 50 to 300 ppm for four surfactant contents. An increase in the inlet concentration generally would enhance the transfer rate of the VOCs from the gas phase to the biofilm (enhancing effect). This phenomenon could explain the fact that more microorganisms were caused to participate in the biodegradation. However, high concentrations of some recalcitrant VOCs may produce inhibitive effects on the metabolic activity of the microbial population (Leson and Winer, 1991). Therefore, the result indicated that the inhibitive effect predominated and the microbial growth rate was inhibited at higher inlet concentration. The result was closely corresponding to the result reported that the higher concentration of toluene and xylenes caused inhibition of microbial growth (Leon et al., 1999; Shim et al., 2005).
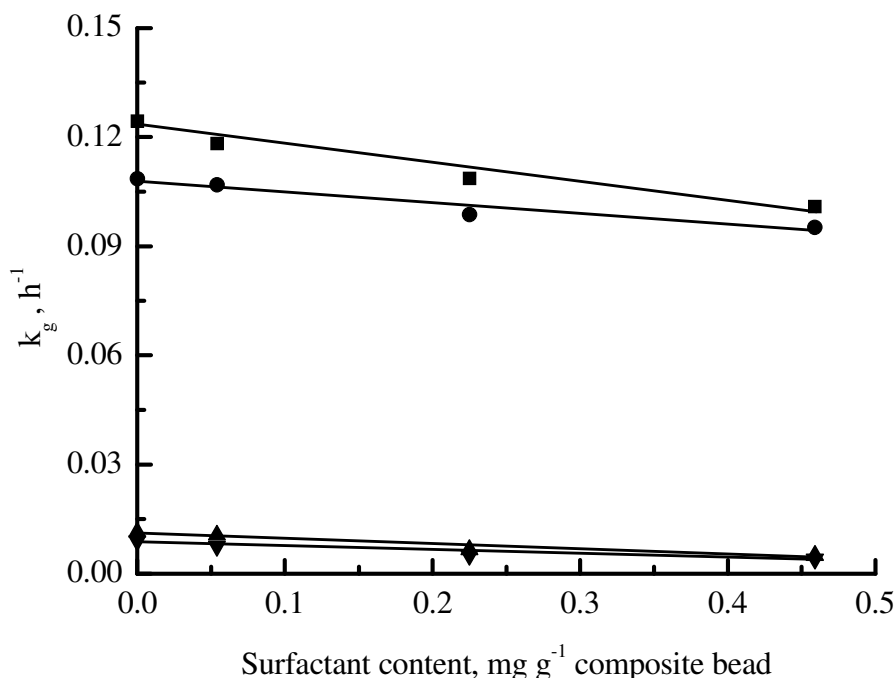
Figure 3. The variations of the k_g values with surfactant content for four inlet concentration: (■) 50 ppm, (●) 100 ppm, (▲) 200 ppm, (▼) 300 ppm.

Biochemical reaction process

In the stationary phase, the population of viable cells was at a relatively constant value. A biofilter modeling that described steady state elimination of individual carbon energy substrates was proposed by Ottengraf. Three basic situations of Ottengraf's model was first-order kinetics, zero-order kinetics with reaction limitation and zero-order kinetics with diffusion limitation (Ottengraf, 1986). The corresponding equations expressed the rates of biochemical reaction for each situation as follows:

First-order kinetic

$$\ln(C/C_0) = - k_1 \theta \qquad (4)$$

Zero-order kinetic with reaction limitation

$$C_0 - C = k_0 \theta \qquad (5)$$

Zero-order kinetic with diffusion limitation

$$C = C_0 [1 - \theta (a k_0 D_e / 2mC_0 \delta)^{1/2}]^2 \qquad (6)$$

where a is the interfacial area per unit volume, D_e is the effective diffusion coefficient, m is the distribution co-efficient of the component, θ is the empty bed residence time (EBRT), δ is the biofilm thickness, and k_1 and k_0 are the rate coefficients of first-order kinetic and zero-order with reaction limitation, respectively. However, for conve-nience of use, it is necessary to define a new parameter, $k_d = (ak_0D_e/2mC_0\delta)^{1/2}$. It can be seen that k_d is a function of the operating conditions of the biofilter system, and k_d is constant under steady-state conditions (Yang and Allen, 1994). Therefore, Eq. 6 can be rewritten as

$$1 - (C/C_0)^{1/2} = k_d \theta \qquad (7)$$

Where k_d is the rate coefficient of zero-order kinetic with diffusion limitation.

The substrate utilization rate by microbial was generally expressed by the Michaeilis-Menten relationship. Under the state of microbial population does not change with time, three possible situations may be encountered in a biochemical reaction system (Yang and Allen, 1994):

Situation 1: if the substrate concentration was very low ($K_s >> C_0$), the reaction rate expression could be simpli-fied to first-order kinetic.
Situation 2: if the substrate concentration was very high ($K_s << C_0$), the reaction rate expression could be simpli-fied to zero-order kinetic.
Situation 3: if the substrate concentration C_0 was compa-rable with K_s, the reaction rate expression could not be simplified and it was followed fractional-order kinetic, and the Ottengraf's diffusion limiting model was found to be
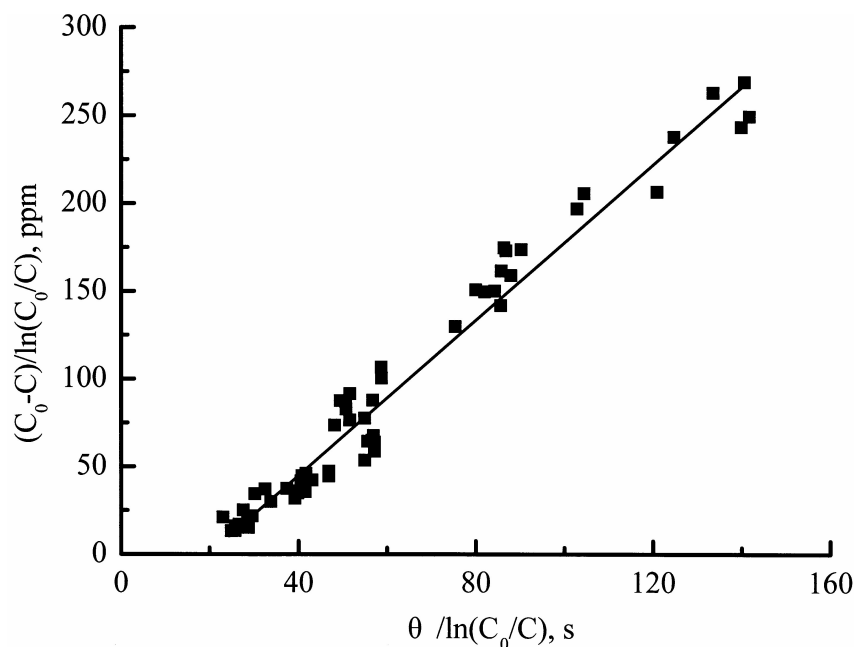
Figure 4. Plot of $(C_0-C)/\ln(C_0/C)$ versus $\theta/\ln(C_0/C)$ at surfactant content of 0.225 mg g^{-1} composite bead.

the most approximate expression.

In order to verify the biochemical reaction kinetic model, assume there was a plug air flow in the biofilter column and the following equation was derived from the Michaelis-Menten equation (Valsaraj, 1995).

$$(C_0-C)/\ln(C_0/C) = V_m (\theta/\ln(C_0/C)) - K_s \quad (8)$$

Where K_s is half-saturation constant and V_m is maximum reaction rate. A plot of $(C_0-C)/\ln(C_0/C)$ versus $\theta/\ln(C_0/C)$ should correspond to a straight line, and K_s and V_m can be determined. The plot of $(C_0-C)/\ln(C_0/C)$ versus $\theta/\ln(C_0/C)$ for four surfactant contents is shown in Figure 4 (only the surfactant content of 0.225 mg g^{-1} composite bead is shown). The calculated K_s for surfactant content 0, 0.054, 0.225 and 0.459 mg g^{-1} composite bead were 24.02, 5.91, 43.55 and 31.18 ppm, respectively. The calculated V_m for surfactant content 0, 0.054, 0.225 and 0.459 mg g^{-1} composite bead were 2.24, 1.59, 2.24 and 1.83 ppm s^{-1}, respectively. The ratio of C_0/K_s for surfactant content 0, 0.054, 0.225 and 0.459 mg g^{-1} composite bead were 2.08-16.65, 8.46 -67.68, 1.15-9.17 and 1.60-12.83, respectively. The results indicated that the relationship of C_0 and K_s was not corresponding to situation 1 or situation 2, but it corresponds to situation 3 for two compounds. Therefore, zero-order kinetic with diffusion limitation was regarded as the most adequate biochemical reaction kinetic model because the values of C_0 and K_s were comparable for four surfactant contents in this study. Thus, the k_d value at various inlet concentrations and surfactant

content was calculated from the data in Phase III and Eq. 7.

The variations of the k_d values with surfactant content for four inlet concentrations are shown in Figure 5. The k_d value decreased with increasing surfactant content in the surfactant content range from 0 to 0.459 mg g^{-1} composite bead. The result indicated that the biochemical reaction rate was also inhibited at higher surfactant content and the addition surfactant into filter material was unfavorable for VOCs degraded by the microbial. The result was closely corresponding to the result reported that the higher concentration of nonionic surfactant Brij30 or Brij 35, the slower biodegradation rate of phenanthrene in the aqueous phase (Yuan et al., 2000). The slope of the linear profiles in the surfactant content range of 0 to 0.459 mg g^{-1} composite bead for 50, 100, 200 and 300 ppm inlet concentration was 1.43×10^{-2}, 3.03×10^{-3}, 4.17×10^{-3} and 1.92×10^{-3} s^{-1} mg^{-1} g composite bead, respectively. The result indicated that the degree of inhibitive effect was more pronounced at lower inlet concentration.

The k_d value in the inlet concentration range of 50 to 100 ppm was much higher than that in the inlet concentration range of 200 to 300 ppm. The result indicated that it also produced toxic to the microorganism in the inlet concentration range of 200 to 300 ppm. The k_d value was decreased with increasing inlet concentration in the inlet concentration range of 50 to 300 ppm for four surfactant contents. The result indicated that the biochemical reaction rate was also inhibited at higher inlet concentration. The result was closely corresponding to the result reported that the pseudo first-order bio-degradation rate constant decreased as the toluene inlet concentration
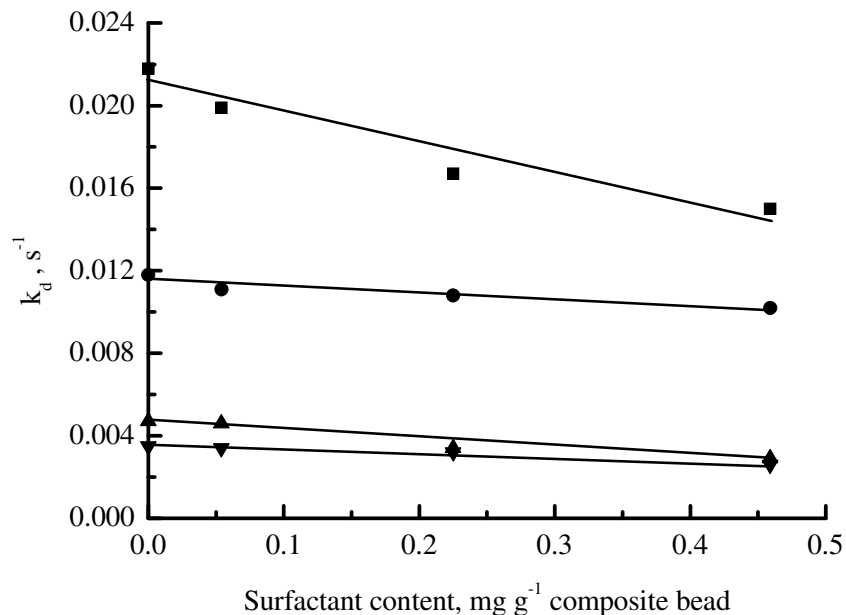
Figure 5. The variations of the k_d values with surfactant content for four inlet concentration: (■) 50 ppm, (●) 100 ppm, (▲) 200 ppm, (▼) 300 ppm.

increased in an acclimated mixed-culture biofilter (Park et al., 2004).

Elimination capacity

Elimination capacity (EC) and load were calculated according to equation presented below: (Deviney et al., 1999)

$$EC = Q (C_0-C)/V \qquad (9)$$

$$Load = Q C_0/V \qquad (10)$$

Where Q is the flow rate of inlet air steam and V is the bed volume of filter material as packed. Under low load conditions, the elimination capacity essentially equals the load, and the system is calculated to have 100% removal efficiency. By increasing the load on a system, a point will be reached where the overall load will exceed the overall elimination capacity, generating removal efficiencies less than 100%. This point is typically called the critical elimination capacity. As the load continues to increase, a maximum overall elimination capacity will eventually be reached. This maximum overall elimination capacity is independent of the contaminant concentration and residence time within a reasonable range of operating conditions (Deviney et al., 1999). The relationship of elimination capacity of biofilter versus load for four surfactant content is shown in Figure 6 (only the surfactant content of 0.225 mg g^{-1} composite bead is shown). It was found that elimination capacity increased and tended towards a constant value with increasing inlet load. The result was

closely corresponding to the result reported in our previous works (Chan and Peng, 2008). The maximum elimination capacity for surfactant content 0, 0.054, 0.225 and 0.459 mg g^{-1} composite bead were 26.12, 24.03, 22.99 and 17.41 g C h^{-1} m^{-3} bed volume, respectively. The result indicated that the maximum elimination capacity decreased with increasing surfactant content in the filter material. Therefore, the addition of surfactant into filter material was unfavorable for toluene degraded by the microbial, as discussed in the earlier section of this paper.

Conclusions

Nonionic surfactant effects on the toluene dissolved in the water phase and biodegradation kinetic behaviors of toluene in an adsorbed nonionic surfactant composite bead biofilter were investigated. The toluene dissolved in the water phase was enhanced by the addition of surfactant into aqueous solution and the enhancing effect by the addition of surfactant in the surfactant concentration less than CMC was more pronounced than that in the surfactant concentration higher than CMC. For the microbial growth process, the microbial growth rate was inhibited at higher surfactant content and it was more pronounced at lower inlet concentration. The microbial growth rate was inhibited at higher inlet concentration. The half-saturation constant K_s values for four surfactant contents were in the range of 5.91 to 43.55 ppm. The maximum reaction rate V_m values for four surfactant contents were in the range of 1.59 to 2.24 ppm s^{-1}. Zero-order kinetic with the diffusion limitation could be regarded as the most adequate bio-chemical reaction
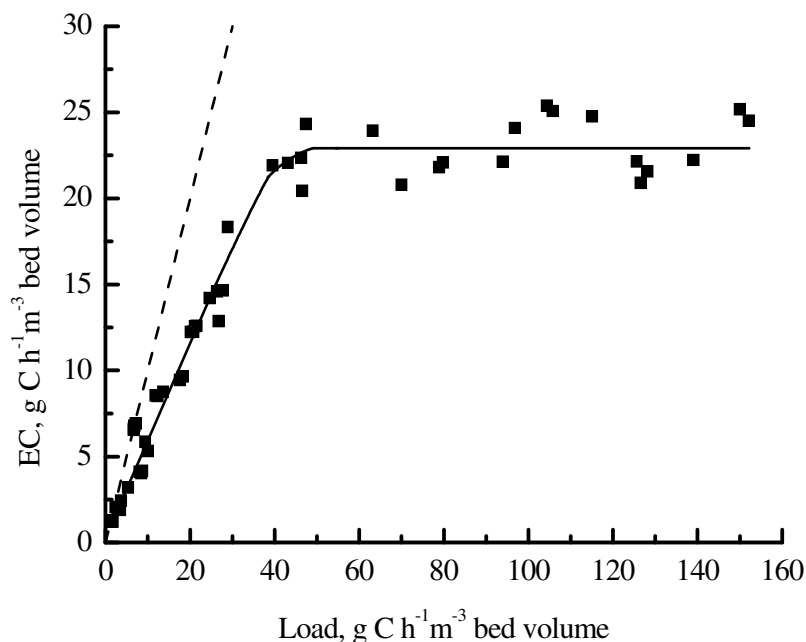
Figure 6. The variations of elimination capacity (EC) with load at the surfactant content of 0.225 mg g^{-1} composite bead: (---) 100% removal.

model. For the biochemical reaction process, the biochemical reaction rate was also inhibited at higher surfactant content and the degree of inhibitive effect was more pronounced at lower inlet concentration. The biochemical reaction rate was also inhibited at higher inlet concentration. The maximum elimination capacity decreased with increasing surfactant content and it was in the range of 17.41 to 26.12 g C h^{-1} m^{-3} bed volume. The addition of surfactant into filter material was unfavorable for toluene degraded by the microbial.

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